ESRF	**Experiment title:** Structural tuning in epitaxial ferromagnetic van der Waals heterostructures probed by grazing incidence diffraction	**Experiment number**: HC-4552
Beamline: BM25B	**Date of experiment**: from: 11.11.2021 to: 16.11.2021	**Date of report**: 01.03.2022
Shifts: 15	**Local contact(s)**: Dr. Juan Rubio Zuazo, Dr. Jesus Lopez Sanchez	*Received at ESRF:*

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Report:

As a direct result out of the granted beamtime we are preparing a manuscript, which is anticipated to be submitted very soon. Thus, we would highly appreciate if you will treat the contained information confidentially. Here we concentrate exclusively on (yet preliminary) results based on the ESRF beamtime without contexting this work into a broader view applying further methods.

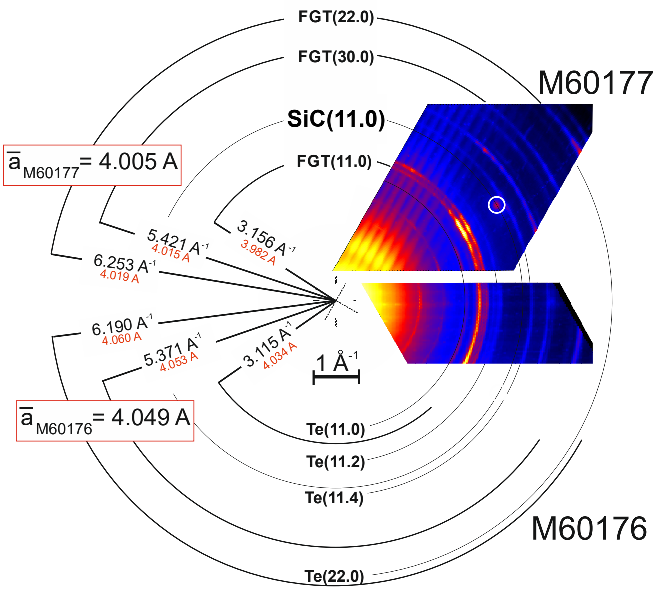

Figure 1: Grazing incidence diffraction in-plane map for two samples containing $Fe_{5-x}GeTe_2$ layers on SiC(00.1) substrates

Our project aimed at X-ray diffraction studies of large-area van der Waals (vdW) heterostructure films containing two-dimensional (2D) magnetic materials synthesized by vdW epitaxy. Previously we obtained important results on the epitaxial properties of vdW heterostructures combining graphene, hexagonal boron nitride (h-BN), as well as molybdenum disulfide and published the respective results. Now we performed grazing incidence diffraction (GID) experiments to investigate novel, all epitaxial vdW heterosystems formed of the layered ferromagnet $Fe_{5-x}GeTe_2$ (FGT) with different Fe content and graphene. This study was crucial for establishing a detailed correlation between structure and magnetism in these material systems. Specifically, we have probed elastic strain and lattice parameters in the ferromagnetic monolayers with different Fe composition, as well as their epitaxial relationship with graphene. Such properties are anticipated to directly influence ferromagnetism in FGT and proximity-induced ferromagnetic exchange interaction in graphene.

We used GID to obtain precise information about strain and epitaxial relationships in different FGT/ graphene stacks synthesized by molecular beam epitaxy under various conditions and possessing different properties. Fig.1 gives an example of two samples with nominally different Fe content (Fe5GeTe2 vs. Fe4GeTe2). Very clearly we could probe slightly different in-plane lattice parameters as shown by sections in reciprocal space parallel to the surface.

Therefor we have prepared large-area FGT films possessing different Fe concentrations and thus distinct phases, spanning the whole range from Fe3GeTe2 to Fe5GeTe2. We also tried different thicknesses of the FGT films from about 8 nm (equivalent to 10 atomic layers) down to a monolayer (not shown here). In-situ capping with amorphous Te or Sb has been performed in-situ to prevent surface oxidation. As substrates we have use high-quality epitaxial graphene on SiC(0001) [and sapphire], which was previously characterized via GID at ESRF. The knowledge obtained in our recent experiment will be closely correlated with results obtained from magneto-optical and magneto-transport measurements (not shown here) performed for twin samples at our institute. This will allow us explore in detail modulations in magnetism that will emerge from precise variations in the crystal structure.

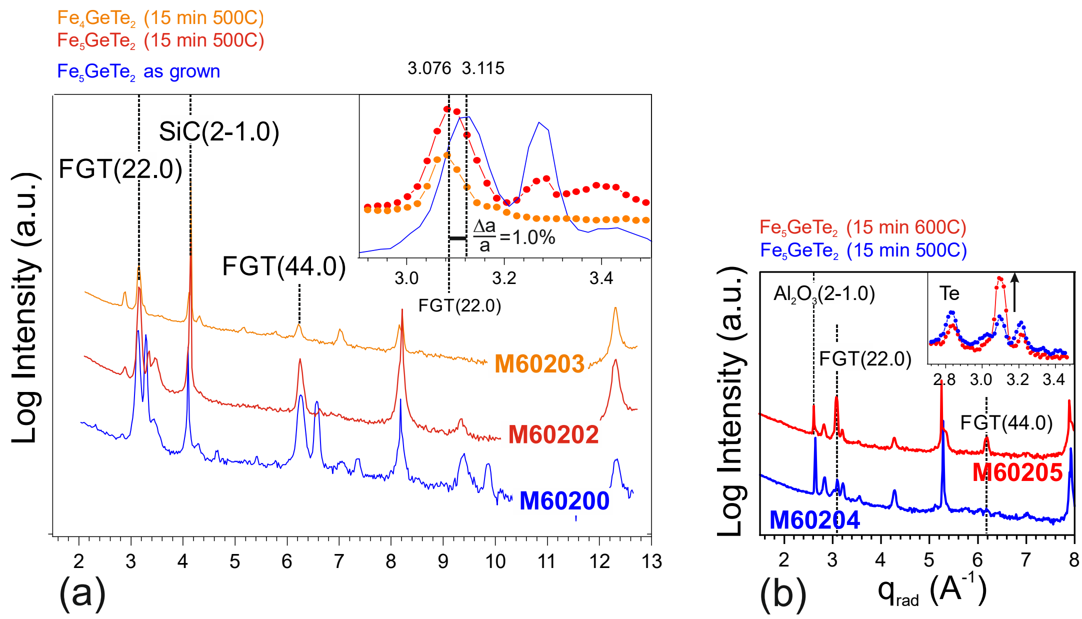

Figure 2: Grazing incidence diffraction in-plane scans for FGT samples containing Fe5GeTe2 (as grown and annealed) and Fe4GeTe2 layers on SiC(00.1) (a) and Al2O3(00.1) substrates (b) demonstrating the effect of thermal annealing.

Further on we will present here first results on the effect of annealing the FGT layers. Fig.2(a) depicts various radial in-plane scans along the SiC[2-1.0] direction for three different samples, namely Fe5GeTe2 (as-grown and annealed) as well as another annealed one with smaller Fe content (Fe4GeTe2). Having a closer look at the FGT(22.0) in-plane peak one can recognize a considerable degree of relaxation of about 1.0 % upon annealing. Thus, we anticipate an improved crystal quality through the annealing step, which happens independent on the particular Fe content. This observation is supported for FGT layers on sapphire substrates as well, see fig.2(b). Similar to the left hand side graph radial scans along the Al2O3[2-1.0] direction are depicted, which intersect the FGT(22.0) reflection as well. The two curves have been taken for layers with same Fe content but annealed at 500 and 600°C, respectively. Obviously the increased annealing temperature of 600°C yields an even larger amount of oriented FGT (increased peak intensity at the red curve).

We may note that the data evaluation is still in progress. But we are rather confident that along with the already performed magneto-optical and magneto-transport measurements we are rather close to submit our results.